



(19) Europäisches Patentamt
European Patent Office
Office européen des brevets



(11) Publication number : 0 662 381 A2

(12)

EUROPEAN PATENT APPLICATION

(21) Application number : 94309859.0

(51) Int. Cl.⁶ : B29C 45/00, C08L 67/00,
C08L 77/12, C09K 19/38,
// (C08L67/00, 23:02),
(C08L77/12, 23:02)

(22) Date of filing : 28.12.94

(30) Priority : 28.12.93 JP 338096/93

(43) Date of publication of application :
12.07.95 Bulletin 95/28

(84) Designated Contracting States :
DE FR GB

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(54) Shaped article of liquid crystalline polymer.

(57) A liquid crystalline polymer contains (A) 100 parts by weight of a liquid crystalline polyester and/or liquid crystal polyesteramide capable of forming an anisotropic melt and (B) 0.01 to 2 parts of an olefinic polymer. The composition is especially suitable for moulding therefrom shaped articles by a moulding process in which at least two flowing molten resin portions of the composition unite to form a weld. Thus, using such a composition it is possible to produce a moulding having such a weld in which the weld has a weld strength retention of 15% to 100%, so that the shaped article has excellent heat stability, mechanical properties, dimensional stability and mold releasability and is suitable for various application uses such as in electrical and electronic devices, accurate machine devices, office related devices and automobile parts.

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The present invention relates to a shaped article of a liquid crystalline polymer having excellent mold releasability from a die when molding, having excellent thermal and chemical stability without generating decomposition gases etc., having a weld with excellently balanced thermal stability such that at increased temperatures the moulding exhibits a low decrease in strength of the weld, mechanical properties and moldability and having excellent dimensional stability.

The need for plastic materials having high performance has been growing in recent years. Numerous polymers having various new types of performance have been developed and marketed. However, optical anisotropic liquid crystalline polymers characterized by the parallel sequences of molecular chains, have received particular attention due to their superior flowability, thermal resistance, mechanical properties, and mechanical stability. Examples of such polymers capable of forming an anisotropic melt include, for example, a liquid crystal polyester obtained by copolymerizing polyethyleneterephthalate with p-hydroxybenzoic acid (Japanese Unexamined Patent Publication No. 49-72393), a liquid crystalline polyester obtained by copolymerizing p-hydroxybenzoic acid and 6-hydroxy-2-naphthoic acid (Japanese Unexamined Patent Publication No. 54-77691), a liquid crystal polyester obtained by copolymerizing 4,4'-dihydroxybiphenyl, terephthalic acid and isophthalic acid with p-hydroxybenzoic acid (Japanese Examined Patent Publication No. 57-24407), a liquid crystalline polyester amide formed from 6-hydroxy-2-naphthoic acid, p-aminophenol and terephthalic acid (Japanese Unexamined Patent Publication No. 57-172921), and a liquid crystalline polyesteramide formed from p-hydroxybenzoic acid, 4,4'-dihydroxybiphenyl, terephthalic acid, p-aminobenzoic acid and polyethylene terephthalate (Japanese Unexamined Patent Publication No. 64-33123).

However, it is also well known in the art that these liquid crystalline polymers have problems including large mechanical anisotropy and dimensional anisotropy. For solving these problems, various attempts have been made. For example, a method wherein a glass fiber is added to a liquid crystalline polymer (Rubber Digest, Vol. 27, No. 8, pp. 7-14, 1975) and a method wherein powder in the form of a plate or flake such as mica, talc or graphite is compounded into a liquid crystalline polymer (Japanese Unexamined Patent Publication No. 63-146959). Thus, the anisotropy is relaxed (or decreased) and simultaneously the mechanical properties, thermal resistance, moldability and dimensional stability are further improved. As a result, the liquid crystalline polymer compositions are used, as engineering plastics, in a wide variety of application fields, such as automobile parts, electrical and electronic parts, precise mechanical parts and office machine parts.

Since liquid crystalline polymers are generally excellent in their flowability, the molding and processing are easy and the liquid crystalline polymers can be used as thin-walled molded products or molded articles having complicated shapes. However, various problems have arisen in the molding of such thin-walled molded products and molded articles having complicated shapes such that the dimensional stability thereof becomes poor due to poor mold releasability and the operation therefore required to remove the molded articles from the mold. Productivity is also decreased.

Generally, as a method for improving the mold releasability, methods for adding various releasing agents such as fatty acid esters, metal salts of fatty acids, fatty acid amides and low molecular weight polyethylene polymers are added to the liquid crystalline polymers are known in the art. In addition Japanese Unexamined Patent Publication No. 2-208035 discloses a method for formulating fatty acid esters into liquid crystalline polyesters and Japanese Unexamined Patent Publication No. 4-120162 discloses a method for adding fatty acid esters composed of polyols (e.g., glycerol and pentaerythritol) and fatty acids having 12 or more carbon atoms.

However, when these methods are applied to liquid crystalline polymers, they cause other problems, due to the high processing temperature of the liquid crystalline polymers, that the releasing agents decompose to generate volatile gases, and therefore, not only is the outside appearance of the molded articles adversely affected, but also the effects of the releasing agents are impaired, whereby the release of the molded article from the die becomes poor and the shape of the molded article is changed (or deformed). Especially in the case of a molded article having a weld (i.e., which is provided by plurality of melted resin flows encountering one another in the mold), the strength at the weld is largely decreased and the strength of the molded articles become poor, so that the molded articles cannot be practically used due to the shortage in the strength of the molded articles, especially in the weld thereof.

Accordingly, the present invention seeks to obviate the above-mentioned problems and to provide a shaped article of a liquid crystalline resin, formed by a moulding process in which at least two flowing molten resin portions unite during the molding to form a weld, which has (a) excellent mold releasability from a mold when molded, (b) good thermal and chemical stability without causing deformation of the molded articles and without generating decomposed gases, (c) only a small decrease in the strength of the weld and (d) despite the presence of the weld, balanced excellent properties of thermal resistance, mechanical properties and moldability, and excellent dimensional stability.

The present invention also seeks to provide shaped articles, e.g., three-dimensional molded articles, sheets, vessels, pipes, of liquid crystalline polymer useful in various fields of application, including electrical

and electronic parts represented by various gears, cases sensors, LEP lamps, connectors, sockets, resistors, relay case switches, coil bobbins, condensers, variable condensers, optical pickups, oscillators, various types of terminal boards, transformers, plugs, printed circuit boards, tuners, speakers, microphones, headphones, small motors, magnetic head bases, power modules, semiconductors, FDD carriages, FDD chassis motor brush holders, parabolic antennae and computer parts; home appliance and electrical office product parts represented by VTR parts, television parts, irons, hair dryers, rice cooker parts, microwave oven parts, audio parts, audio equipment parts such as those for audio laser disks and compact disks, lighting parts, refrigerator parts, air conditioner parts, typewriter parts and word processor parts; mechanical parts represented by office computer parts, telephone parts, facsimile parts, copier parts, cleaning tools, oilless bearings, stern bearings, water bearings, motor parts, lighters and typewriters; optical equipment and precision machinery parts represented by microscopes, binoculars, cameras and clocks; and, automotive and vehicular parts such as alternator terminals, alternator connectors, IC regulators, potentiometer bases, various valves such as exhaust gas valves, various fuel, exhaust and air intake pipes, air intake nozzle snorkels, intake manifolds, fuel pumps, engine coolant joints, carburetor main bodies, carburetor spacers, exhaust gas sensors, coolant sensors, oil temperature sensors, brake pad wear sensors, throttle position sensors, crankshaft position sensors, air flow meters, brake pad wear sensors, air conditioner thermostat bases, heater air flow control valves, radiator motor brush holders, water pump impellers, turbine vanes, wiper motor parts, distributors, starter switches, starter relays, transmission wire harnesses, window washer nozzles, air conditioner panel switch plates, fuel system solenoid valve coils, fuse connectors, horn terminals, electrical equipment insulating plates, step motor rotors, lamp sockets, lamp reflectors, lamp housings, brake pistons, solenoid bobbins, engine oil filters and ignition device cases.

The following provides a description of the constitution of such shaped articles.

According to a first aspect, the present invention provides a composition comprising (A) 100 parts by weight of at least one liquid crystalline polymer selected from the group consisting of liquid crystalline polyesters and liquid crystalline polyester amides capable of forming an anisotropic melt; and (B) 0.01 to 2 parts by weight of at least one olefinic polymer having a weight-average molecular weight of 10,000 to 600,000 and selected from the group consisting of (a) polyethylene, (b) polypropylene, (c) copolymers of ethylene and α -olefin having 3 or more carbon atoms, (d) copolymers of propylene and α -olefin having 4 or more carbon atoms, (e) copolymers of ethylene, α -olefin having 3 or more carbon atoms and a non-conjugated diene and (f) copolymers of propylene, α -olefin having 4 or more carbon atoms and a non-conjugated diene.

According to a second aspect, the invention provides a shaped article of a liquid crystalline polymer having a weld, which is provided by plural melted resin flows encountering in the mold, obtained by melt molding the above composition.

According to a third aspect, the invention provides such a shaped article having a weld strength retention 15% to 100% obtained by the following formula (1), based upon flexural strengths determined from a weld shaped article (X) having a weld, which is provided by plural melted resin flows encountering in the mold, at substantially the center portion thereof and a non-weld shaped article (Y) having no weld, both obtained by injection molding the composition under the following conditions:

$$\text{Weld strength retention (\%)} = (\text{Flexural strength of shaped article (X) having weld}/\text{Flexural strength of shaped article (Y) having no weld}) \times 100 \quad (1)$$

Injection molding condition:

Cylinder temperature: Melting point ($^{\circ}\text{C}$) of liquid crystalline polymer + 10°C

Mold temperature: 90°C

Size of shaped article: 6.0 mm (width) \times 127 mm (length) \times 1 mm (thickness).

In accordance with the present invention, there is also provided the above-mentioned shaped article having a weld with a weld strength retention of the weld of 20% to 100%.

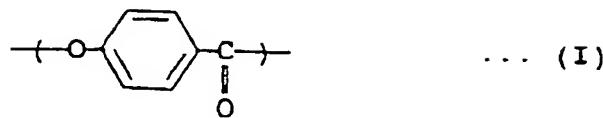
In accordance with the present invention, there is further provided a composition and shaped article in accordance with any of the above aspects, wherein the liquid crystalline polymer (A) is at least one polymer selected from the group consisting of fully aromatic liquid crystalline polyesters, fully aromatic liquid crystalline polyesteramides, liquid crystalline polyesters having an ethylenedioxy unit, and liquid crystalline polyesteramides having an ethylenedioxy unit.

In a preferred composition and shaped article according to any of the above aspects, the liquid crystalline polyester (A) comprises at least one liquid polyester components each said component containing a combination of structural units selected from (i)(I), (III) and (IV), (ii)(I), (II) and (IV) and (iii)(I), (II), (III) and (IV).

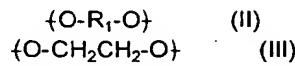
In accordance with the present invention, there is still further provided the above-mentioned composition and shaped article according to any of the above aspects, wherein the liquid crystalline polyester (A) is at least one liquid crystalline polyester composed of the following structural units (I), (III) and (IV), liquid crystalline polyesters composed of the following structural units (I), (II) and (IV), and liquid crystalline polyesters having

the following structural units (I), (II), (III) and (IV):

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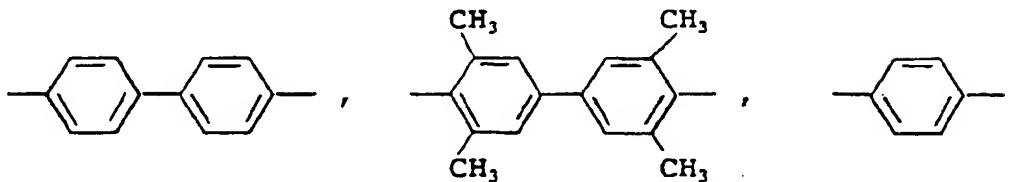


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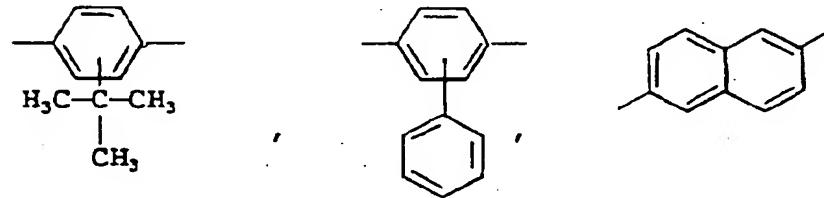
wherein R_1 represents at least one group selected from the group consisting of:

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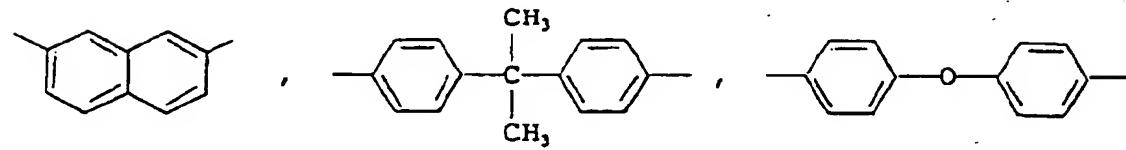
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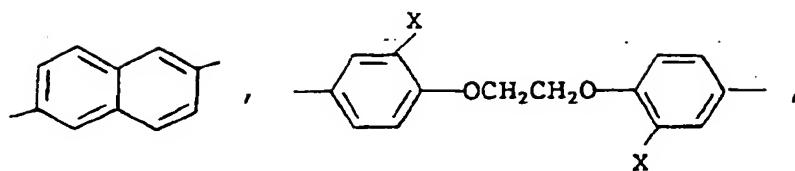
and R_2 represents at least one group selected from the group consisting of:

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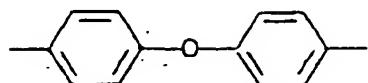
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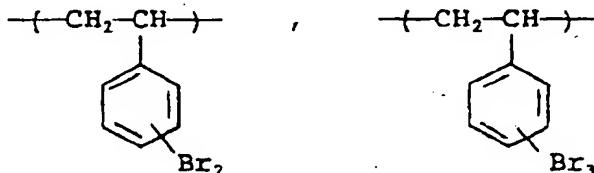
15 wherein X represents a hydrogen atom or chlorine atom, and the total number of moles of structural unit (II) and structural (III) are substantially equal to the total of the number of moles of structural unit (IV).

In accordance with the present invention, there is still further provided any of the above-mentioned aspects, wherein the composition further contains:

- 20 (1) 400 parts by weight or less of a filler, based upon 100 parts by weight of the liquid crystalline polymer (A); and/or
- (2) 0.01 to 10 parts by weight of carbon black, based upon 100 parts by weight of the liquid crystalline polymer (A); and/or
- (3) 0.5 to 60 parts by weight of an organic bromine compound based upon 100 parts by weight of the liquid crystalline polymer (A).

25 The organic bromine compound is preferably a poly(brominated styrene) having a weight average molecular weight of 1×10^3 to 120×10^4 and having at least one of the following structural units obtained from brominated styrene monomer as the major structural component.

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Preferred embodiments of the invention will now be described in more detail with reference to the accompanying drawings in which Figs. 1 and 2 schematically illustrate perspective views of a weld molded article (X) and a box-type molded article obtained in Example 1, respectively.

PREFERRED EMBODIMENTS OF THE INVENTION

In a composition embodying the present invention, as the component (A), at least one liquid crystalline polymer selected from liquid crystalline polyesters, and including liquid crystalline polyesteramides, capable of forming an anisotropic melt is used.

The liquid crystalline polyesters and the liquid crystalline polyesteramides capable of forming an anisotropic melt referred to herein may be any liquid crystalline polyester which forms an anisotropic melt comprising structural units selected from structural units such as an aromatic oxycarbonyl unit, an aromatic dioxy unit, an aromatic dicarbonyl unit and an ethylenedioxy unit, or liquid crystalline polyesteramide capable of forming an anisotropic melt comprising structural units such as the above-mentioned structural units, an aromatic iminocarbonyl unit, an aromatic diimino unit and an aromatic iminoxy unit.

The liquid crystalline polyester and/or polyesteramide capable of forming an anisotropic melt used in the present invention may be a fully aromatic thermotropic polyester, preferably one having a naphthalene ring, a fully aromatic thermotropic polyesteramide, preferably one having a naphthalene ring, a liquid crystalline polyester having ethylenedioxy units, or a liquid crystalline polyesteramide having ethylenedioxy amide units capable of forming an anisotropic molten phase.

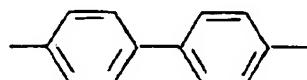
Preferable examples of liquid crystalline polyesters include liquid crystalline polyesters comprising the

structural units of (I), (III) and (IV), (I), (II) and (IV) or (I), (II), (III) and (IV). Among these, the use of a liquid crystalline polyester having structural units (I), (II) and (IV) or having structural units (I), (II), (III) and (IV) is preferable.

Such units may be formed by reaction of an alcohol, acid or amino group or any reactive derivative thereof, but for convenience reference is made below only to the free alcohol, acid or amine.

Structural unit (I) is a structural unit of a polyester derived from p-hydroxybenzoic acid. Structural unit (II) represents a structural unit derived from an aromatic dihydroxy compound selected from 4,4'-dihydroxybiphenyl, 3,3'5,5'-tetramethyl-4,4'-dihydroxyphenyl, hydroquinone, t-butylhydroquinone, phenylhydroquinone, 2,6-dihydroxynaphthalene, 2,7-dihydroxynaphthalene, 2,2-bis(4-hydroxyphenyl)propane and 4,4'-dihydroxyphenylether. Structural unit (III) represents a structural unit formed from ethyleneglycol. Structural unit (IV) represents a structural unit formed from an aromatic dicarboxylic acid selected from terephthalic acid, isophthalic acid, 4,4'-diphenyldicarboxylic acid, 2,6-naphthalenedicarboxylic acid, 1,2-bis(phenoxy)ethane-4,4'-dicarboxylic acid, 1,2-bis(2-chlorophenoxy)ethane-4,4'-dicarboxylic acid and 4,4'-diphenylether dicarboxylic acid.

Among these, especially preferred compounds are those having, as R₁:



and having, as R₂:



The liquid crystalline polymers preferably used in the present invention are liquid crystalline polyesters comprising the above-mentioned structural units (I), (II) and (IV) or (I), (II), (III) and (IV), and the copolymerized amounts of the above-mentioned structural units (I), (II), (III) and (IV) are not limited. However, the following copolymerized amounts are preferable having regard to the flowability property of the composition.

Namely, in the case of those containing the above-mentioned structural unit (III), the molar fraction of the total of the structural units (I) and (II) to the total of the above-mentioned structural units (I), (II) and (III) is preferably 60 to 95 mol%, particularly preferably 75 to 93 mol% having regard to the thermal resistance, flame resistance and mechanical properties of the composition. In addition, the molar fraction of the structural unit (III) to the total of the structural units (I), (II) and (III) is preferably 40 to 5 mol%, particularly preferably 25 to 7 mol%.

In addition, the molar fraction ratio (I)/(II) of the structural unit (I) to the structural unit (II) is preferably 75/25 to 95/5, and particularly preferably 78/22 to 93/7 in achieving a balance between the thermal resistance and the flowability. Furthermore, it is preferable that, the total number of moles of the structural unit (II) and the structural unit (III) is substantially equal to the number of moles of the structural unit (IV). The term "substantially" used herein means that, if desired, the number of either of the carboxy end group or the hydroxyl end group in the end group of the polyester can be made larger, but in such a case, the total number of moles of the structural units (II) and (III) is not completely equal to the number of moles of the structural unit (IV).

On the other hand, in the case of those not containing the above-mentioned structural unit (III), the molar fraction of the structural unit (I) to the total of the structural units (I) and (II) is preferably 40 to 90 mol%, and particularly preferably 60 to 88 mol%. It is preferable that the number of moles of the structural unit (II) is substantially equal to the number of moles of the structural unit (IV). In this case, the meanings of the term "substantial" is the same as mentioned in the above case including the unit (III). This is the same as in the case of the liquid crystalline polyesters having the structural units of (I), (III) and (IV).

Furthermore, when the above-mentioned liquid crystalline polyester particularly preferably used in the present invention is a polycondensate, at least one of aromatic dicarboxylic acids such as 3,3'-diphenyldicarboxylic acid and 2,2'-diphenyldicarboxylic acid, aliphatic dicarboxylic acids such as adipic acid, azelaic acid, sebacic acid and dodecanedionic acid, alicyclic dicarboxylic acids such as hexahydroterephthalic acid, aromatic diols such as chlorohydroquinone, methylhydroquinone, 4,4'-dihydroxydiphenylsulfone, 4,4'-dihydroxydiphenylsulfide and 4,4'-dihydroxybenzophenone, aliphatic and alicyclic diols such as 1,4-butanediol, 1,6-hexanediol, neopentylglycol, 1,4-cyclohexanediol and 1,4-cyclohexanedimethanol, aromatic hydroxycarboxylic acids such as m-hydroxybenzoic acid and 2,6-hydroxynaphthoic acid, and p-aminophenol and p-aminobenzoic

acid can be additionally used, other than the above-mentioned components forming the structural units (I) to (IV) within a range of low proportions to an extent that does not unduly impair the desired properties provided by a composition of the present invention.

In addition to the above structural units (I) to (IV), as the liquid crystalline polyester amides, a polyester amide capable of forming an anisotropic melt phase and containing a p-imino phenoxy unit formed from p-aminophenol can be used.

There are no particular limitations as to the process for producing the above-mentioned liquid crystalline polyesters preferably used in the present invention and the liquid crystalline polyesters and polyesteramides can be produced according to any known polyester or polyesteramide condensation polymerization methods.

For example, those not containing the above-mentioned structure unit (III) are preferably produced by the following methods (1) and (2), and

those containing the structural unit (III) are preferably produced by the following method (3).

(1) A production process using a condensation polymerization with the removal of acetic acid from a diacrylated product of p-acetoxybenzoic acid, an aromatic dihydroxy compound such as 4,4'-diacetoxypiphenyl or para-4,4'-diacetoxylbenzene, and an aromatic dicarboxylic acid such as terephthalic acid.

(2) A production process using a condensation polymerization reaction with the removal of acetic acid from the reaction of p-hydroxybenzoic acid and an aromatic dihydroxy compound such as 4,4'-dihydroxypiphenyl or hydroxyquinone, and acetic anhydride with an aromatic dicarboxylic acid such as terephthalic acid, followed by acylation of phenolic hydroxyl groups.

(3) A production process according to the above-mentioned (1) or (2) carried out in the presence of a polymer or oligomer such as polyethylene terephthalate, or a bis(β-hydroxyether)ester of an aromatic dicarboxylic acid such as bis(β-hydroxyethyl)terephthalate.

In addition, these condensation polymerization reactions can be carried out without using a catalyst, but a catalyst may be optionally used. Typical examples of such catalysts usable in the condensation polymerization reaction include metal compounds such as stannous acetate, tetrabutyltitanate, potassium acetate, sodium acetate, antimony trioxide and magnesium metal.

The logarithmic viscosity (or inherent viscosity) of the liquid crystal polymer (A) according to the present invention can be measured in pentafluorophenol. It is preferable that the value measured at 60°C at a concentration of 0.1 g/dl is at least 0.5 dl/g, and particularly in the case of those containing the above-mentioned structural unit (III), the value of 1.0 to 3.0 dl/g is preferable. On the other hand, in the case of those not containing the above-mentioned structural unit (III), the logarithmic viscosity (or inherent viscosity) value of 2.0 to 10.0 dl/g is preferable.

In addition, the melt viscosity of the liquid crystal polymer (A) usable in the present invention is preferably 10 to 20,000 poise, and more preferably 20 to 10,000 poise.

Furthermore, the above-mentioned melt viscosity refers to the value measured at a temperature 10°C higher than the melting point (T_m) (°C) of the liquid crystal resin (A) (i.e. $T_m + 10^\circ\text{C}$) using a Koka type flow tester under the conditions of a shear rate of 1,000 (1/sec) and a nozzle size of 0.5 mm in diameter by 10 mm in length.

The melting point (T_m) (°C) referred herein represents the endothermic peak temperature which is observed when the measurement is carried out at a temperature-elevating rate of 20°C/min by using a differential scanning calorimeter, i.e., T_{m_2} , described hereinafter.

For the differential scanning calorimetric measurement described above, a polymer obtained by polymerization is heated from room temperature to a temperature exceeding the melting point at a temperature-elevating rate of 20°C/min, and the observed endothermic peak temperature (hereinafter referred to as " T_{m_1} ") is measured. After the measurement of T_{m_1} , the polymer is maintained at a temperature of $T_{m_1} + 20^\circ\text{C}$ for 5 minutes, followed by cooling once down to room temperature at a temperature-dropping rate of 20°C/min, and the temperature is then elevated at a rate of 20°C/min. The endothermic peak temperature (hereinafter referred to as " T_{m_2} ") measured at this second run is measured.

The olefinic polymer usable in the present invention is present in the composition in an amount of from 0.01 to 2, preferably 0.05 to 1.5 parts, inclusive, by weight per 100 parts of polyester and/or polyesteramide and is at least one olefinic polymer selected from the group consisting of (a) polyethylene, (b) polypropylene, (c) copolymers of ethylene and α-olefin having 3 or more carbon atoms, preferably 3 to 20 carbon atoms, (d) copolymers of propylene and α-olefin having 4 or more carbon atoms, preferably 4 to 20 carbon atoms, (e) copolymers of ethylene, α-olefin having 3 or more carbon atoms, preferably 3 to 20 carbon atoms, and a non-conjugated diene and (f) copolymers of propylene, α-olefin having 4 or more carbon atoms, preferably 4 to 20 carbon atoms, and a non-conjugated diene.

Examples of the preferable α-olefins having 3 or more carbon atoms are propylene, butene-1, pentene-1, 3-methylpentene-1, octacene-1. These may be used alone or in any mixture thereof. The use of propylene and butene-1 is further preferable.

Examples of the preferable α -olefins having 4 or more carbon atoms are those having 3 or more carbon atoms exemplified above, other than propylene. These may be used alone or in any mixture thereof.

Examples of the preferable non-conjugated dienes are 5-ethylidene-2-norbornene, dicydopentadiene and 1,4-hexadiene. Again, these non-conjugated dienes may be used alone or in any mixture thereof.

5 The ratio of ethylene/ α -olefin having 3 or more carbon atoms in the copolymer thereof according to the present invention is preferably 40/60 - 99/1 (mole ratio), more preferably 70/30 - 95/5 (mole ratio). The ratio of propylene α -olefin having 4 or more carbon atoms in the copolymer thereof is preferably 40/60 - 99/1 (mole ratio), more preferably, 70/30 - 95/5 (mole ratio).

10 The ethylene content of the ethylene/C₃ or more α -olefin/non-conjugated diene copolymer is preferably 5 - 96.9 mole%, more preferably 30 - 84.5 mole%. The amount of α -olefin having 3 or more carbon atoms is preferably 3 - 80 mole%, more preferably 15 - 60 mole%, and the amount of non-conjugated diene is preferably 0.1 - 15 mole%, more preferably 0.5 - 10 mole%. Furthermore, the propylene content of the propylene/ α -olefin having C₄ or more carbon atoms/non-conjugated diene is 5 - 96.9 mole% more preferably 30 - 84.5 mole% and the content of α -olefin having 4 or more carbon atoms is preferably 3 - 80 mole%, more preferably 15 - 60 mole%, and the content of the non-conjugated diene is preferably 0.1 to 15 mole%, more preferably 0.5 to 10 mole%.

15 Examples of the above-mentioned copolymers are ethylene/propylene copolymer, ethylene/butene-1 copolymer, ethylene/pentene-1 copolymer, ethylene/propylene/butene-1 copolymer, propylene/pentene-1 copolymer, propylene/butene-1 copolymer, propylene/pentene-1 copolymer, propylene/butene-1 copolymer, ethylene/propylene/5-ethylidene-2-norbornene copolymer, ethylene/propylene/1,4-hexadiene copolymer, propylene/butene-1/1,4-hexadiene copolymer, ethylene/propylene/dicyclopentadiene copolymer, etc. Among these, the use of ethylene/propylene copolymer and ethylene/butene-1 copolymer is especially preferable due to the excellent thermal stability thereof. These olefinic copolymers may be used alone or in any mixture thereof. In addition, it is preferable from the viewpoint of the flowability of the resultant composition that a comonomer having an epoxy and/or carboxylic acid group is not copolymerized therein.

20 As mentioned above, according to the present invention, the liquid crystalline polymer composition should have a retention rate of a weld strength of 15% to 100%, preferably 20% to 100%, particularly, preferably 25% to 100%, determined by the following method. When the retention rate of a weld strength is less than 15%, the strength of the weld of the liquid crystalline polymer shaped article is low and is not practically acceptable.

25 Although the upper limit is preferably 100%, the retention rate of up to 60% is particularly acceptable.

30 The retention rate of the weld strength can be determined according to the present invention as follows.
 (1) The test piece (i.e., molded article) is prepared as follows.

A. Test piece

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The test piece is prepared by an injection molding under the following conditions

Cylinder temperature: (Melting point (°C) of liquid crystalline polymer (A) + 10)°C

Mold temperature: 90°C

Size of shaped article: 6.0 mm (width) × 127 mm (length) × 1 mm (thickness)

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More specifically, weld molded articles (X) having a weld, which is provided by plural melted resin flows encountering in the mold, around the center portion of 6.0 mmW × 127 mmL × 1 mmT are prepared by injection molding, using a mold provided with gates having a width of 3 mm at the positions of 3 mm from the both edge portions in the lengthwise direction, under the conditions of a cylinder temperature of an injection molding machine of (the melting point of liquid crystalline polymer (A) + 10)(°C) and a mold temperature of 90°C. Furthermore, non-weld molded articles (Y) having no weld is prepared from one gate by injection molding under the same conditions (i.e., a cylinder temperature of an injection molding machine of (the melting point of the liquid crystalline polymer (A) + 10)(°C) and a mold temperature of 90°C).

Note: As an error for the condition setting in an injection molding machine, the following ranges are usually allowable.

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Cylinder temperature: (Melting point of liquid crystalline polymer (A) + 10)(°C) ± 8°C

Mold temperature: 90°C ± 10°C

Size of molded article: 6.0 mm ± 0.30 mm (width) × 127 mm ± 3 mm (length) × 1 mm ± 0.2 mm (thickness)

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When the melting point of the liquid crystalline polymer used in the evaluation is difficult to determine, the cylinder temperature can be substituted with a temperature of (liquid crystal initiation temperature + 40)(°C) and the allowable error range is (liquid crystal initiation temperature + 40)(°C) ± 8°C.

Fig. 1 shows a perspective view of the weld molded article (X), obtained by injection molding (1) having a size of 127 mm (length L₁) × 6.0 mm (width W₁) × 1 mm (thickness t₁) wherein the two gate positions 2 are

provided at the position of 3 mm from the both edge portions in the lengthwise direction.

(2) Determination method of retention rate of weld strength

The test pieces obtained above are maintained at a temperature of $23^{\circ}\text{C} \pm 2^{\circ}\text{C}$ (i.e., room temperature) and a relative humidity of $50\% \pm 5\%$ for 15 hours.

Then, the flexural strengths of, the weld molded article (X) and non-weld molded article (Y) are determined under the conditions of a spun distance of 20 mm and a strain rate of 0.7 mm/min. The retention rate of weld strength is calculated by substituting the determined value of the flexural strength to the equation (1). Thus, the determination of the retention rate of weld strength is determined from the following equation (1), based upon flexural strengths determined from a weld molded article (X) having a weld at substantially the center portion thereof and a non-weld molded article (Y) having no weld:

$$\text{Retention rate of weld strength (\%)} = (\text{Flexural strength of molded article (X) having weld}) / (\text{Flexural strength of molded article (Y) having no weld}) \times 100 \quad (1)$$

The molded article of the liquid crystalline polymer according to the present invention can be produced by melt molding the above-mentioned liquid crystalline composition. As to the melt molding method, there is no specific limitation and various molding methods including injection molding, extrusion molding and blow molding can be used. Among these methods, injection molding is preferred. As a preferable injection molding method, it is recommendable to feed the above-mentioned liquid crystalline polymer composition to an in-line type injection molding machine set at a temperature within the range of $T_m - 30^{\circ}\text{C}$ to $T_m + 50^{\circ}\text{C}$ (Note: T_m = melting point of the liquid crystalline polymer composition) (or when the melting point is difficult to determine, a temperature within the range of the liquid crystalline initiation temperature + 10°C to the liquid crystalline initiation temperature + 90°C), followed by injection molding to a mold having a shape corresponding to the desired article and set at a mold temperature of about 70°C to about 150°C . Thus, the molded article of liquid crystalline composition having a weld can be molded.

The molded article according to the present invention can be molded, for example, as mentioned above, but it is necessary that the molded article has at least one weld.

The "weld" used herein means a portion which is provided by plural (at least two) melted resin flows encountering one another in the mold. Both resins of respective flows may be the same as or different from one another.

EXAMPLES

Embodiments of the present invention will now be further illustrated by following Examples.

Reference Example 1

A 994 part by weight amount of p-hydroxybenzoic acid, 126 parts by weight of 4,4'-dihydroxybiphenyl, 112 parts by weight of terephthalic acid, 216 parts by weight of polyethylene terephthalate having an inherent viscosity of approximately 0.6 dl/g and 960 parts by weight of acetic anhydride were charged into a reaction vessel equipped with a stirrer and a distillation tube to obtain the desired liquid crystalline polymer (A) by the completion of condensation polymerization. The melting point of the resultant polymer (A) was 314°C and a melt viscosity at 324°C and a shear rate of 1,000/sec was 400 poise.

Reference Example 2

A 994 part by weight amount of p-hydroxybenzoic acid, 222 parts by weight of 4,4'-dihydroxybiphenyl, 147 parts by weight of 2,6-diacetoxynaphthalene, 1078 parts by weight of acetic anhydride and 299 parts by weight of terephthalic acid were charged into a reaction vessel equipped with a stirrer and a distillation tube to obtain the desired polymer (B) by the completion of condensation polymerization.

The melting point (T_m) of this polymer was 336°C and a melt viscosity at a temperature of 346°C and a shear rate of 1,000/sec was 520 poise.

Reference Example 3

A 1296 parts by weight amount of p-acetoxybenzoic acid and 346 parts by weight of polyethylene terephthalate having an inherent viscosity of approximately 0.6 dl/g were charged into a reaction vessel equipped with a stirrer and a distillation tube according to Japanese Unexamined Patent Publication No. 49-72393, followed by condensation polymerization to obtain the desired polymer (C) having a melting point (T_m) of 283°C and a melt viscosity at a temperature of 293°C and a shear rate of 1,000/sec was 1,200 poise.

Reference Example 4

5 A 921 parts by weight amount of p-acetoxybenzoic acid and 435 parts by weight of 6-acetoxy-2-naphthoic
 Unexamined Patent Publication No. 54-77691, followed by condensation polymerization to obtain the desired
 polymer (D) having a melting point (Tm) of 283°C and a melt viscosity at a temperature of 293°C and a shear
 rate of 1,000/sec of 2,000 poise.

Examples 1-4 and Comparative Example 1-5

10 The liquid crystalline polymers obtained in Reference Examples 1-4 were dry blended with olefinic polymers listed in Table 1 in amounts shown in Table 1. Thereafter, the blend was melt kneaded to obtain pellets, using a twin-screw extruder having a diameter of 30 mm in which the cylinder temperature was set to the melting point of each polymer. The resultant pellets were then fed into a Sumitomo Nestal Promat 40/25 Injection Molding Machine (Sumitomo Heavy Machine Industry K.K.) and injection molded into weld molded products (X) having a weld at substantially the center portion thereof and non-weld molded products (Y) having no weld having a size of 6 mmW × 127 mmL × 1 mmT under the conditions of a cylinder temperature of Tm + 10°C and a mold temperature of 90°C.

20 The molded products were brought to and maintained at a temperature of 23 ± 2°C and a relative humidity of 50 ± 5% for 15 hours or more and, then, the flexural strengths of the weld molded product (X) having a weld at substantially the center portion thereof and a non-weld molded product (Y) having no weld were determined at a span distance of 25 mm and a strain rate of 1 mm and the retention rate of the weld strength was obtained by substituting the values of the flexural strengths to the equation (1):

$$25 \quad \text{Retention rate of weld strength (\%)} = (\text{Flexural strength of molded product (X) having weld}) / (\text{Flexural strength of molded product (Y) having no weld}) \times 100 \quad (1)$$

30 Furthermore, the above pellets were fed to a Toshiba 1S55 EPN injection molding machine (Toshiba Kikai Plastic Engineering Co.) to obtain a molded product in the form of a box having a size of 8 mm width × 10 mm height × 100 mm length × 1 mm thickness under the conditions of a cylinder temperature of (Tm of the liquid crystalline polymer +10)(°C) and a mold temperature of 90°C. The box type molded product thus obtained has four partition walls having a thickness of 0.8 mm placed with the equal spacing, which was injection molded at the two gates. When the molded product was ejected, the ejection force (i.e., mold releasing force) was determined, whereby the mold releasability was evaluated.

35 Fig. 2 shows a perspective view of the above box type molded product, wherein the molded product (or article) 3 was injection molded from the gate position 2 provided at the two positions. The size of the box type molded article is 8 mm width (W₂) × 10 mm height (t₂) × 100 mm length (L₂).

40 When the above-mentioned ejection force (i.e., mold releasing force) is smaller, the mold releasability is excellent. Furthermore, the presence or absence of the deformation by the ejection pin when released from the mold was visually observed by the shape of the partition portion of the molded product. This is the measure for dimensional stability. The results are shown in Table 1.

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Table 1

	Liquid crystalline polymer			Olefinic polymer			Retention rate of weld strength (wt. part)	(%)	Performance of box type molded product
	Kind	Amount	Kind	Mw	Amount	Change in shape of partition portion			
Example 1	A	100	PE	4.0×10^5	0.3	22	35	None	
5"	B	100	PE	4.0×10^5	0.3	22	35	"	
"	C	100	PE	4.0×10^5	0.3	16	40	"	
"	D	100	PE	4.0×10^5	0.3	15	42	"	
Comparative Ex.	A	100	-	-	-	23	92	Yes	
10 "	B	100	-	-	-	23	92	"	
"	C	100	-	-	-	18	95	"	
"	D	100	-	-	-	16	95	"	
"	D	100	PE	-	2.04	13	30	"	

15 Box type molded product: 8 mm width x 10 mm height x 100 mm length x 1 mm thickness
with 0.8 mm thick partition

Olefinic polymer:
PE = polyethylene

Examples 5-17 and Comparative Examples 6-12

The liquid crystalline polymers obtained in Reference Examples 1-4 were dry blended with olefinic poly-

mers and the other additives listed in Table 2 in amounts shown in Table 2. Thereafter, the blend was melt kneaded to obtain pellets, using a twin-screw extruder having a diameter of 30 mm in which the cylinder temperature was set to the melting point of each polymer. The resultant pellets were then fed into a Sumitomo Nestal Promat 40/25 Injection Molding Machine (Sumitomo Heavy Machine Industry K.K.) and injection molded into weld molded products (X) having a weld at substantially the center portion thereof and non-weld molded products (Y) having no weld having a size of 6 mm W × 127 mm L × 1 mm T under the conditions of a cylinder temperature of $T_m + 10^\circ\text{C}$ and a mold temperature of 90°C .

The molded products were brought to and maintained at a temperature of $23 \pm 2^\circ\text{C}$ and a relative humidity of $50 \pm 5\%$ for 15 hours or more and, then, the flexural strengths of the weld molded product (X) having a weld at substantially the center portion thereof and a non-weld molded product (Y) having no weld were determined at a spun distance of 25 mm and a strain rate of 1 mm and the retention rate of the weld strength was obtained by substituting the values of the flexural strengths to the above-mentioned equation (1):

Furthermore, the above pellets were fed to a Toshiba 1S55EPN injection molding machine (Toshiba Machine Plastic Engineering Co., Ltd.) to obtain a molded product in the form of a box having a size of 8 mm width × 10 mm height × 100 mm length × 1 mm thickness, as in Example 1 under the conditions of a cylinder temperature of (T_m of the liquid crystalline polymer + 10) ($^\circ\text{C}$) and a mold temperature of 90°C .

As in Example 1, when the molded product was ejected, the ejection force (i.e., mold releasing force) was determined, whereby the mold releasability was evaluated and also the presence or absence of the deformation by the ejection pin when released from the mold was visually observed. Furthermore, the flexural test was carried out under the conditions of a spun distance of 50 mm and a strain rate of 3 mm and the strength generating cracks at the weld was determined. The results are shown in Table 2.

In Table II, the olefine polymers are shown as follows.

PE = polyethylene

PP = polypropylene

PE/P = ethylene/propylene copolymer (mole ratio 83/17)

PE/B1 = ethylene/butene-1 copolymer (mole ratio 90/10)

PE/P/HD = ethylene/propylene/1,4-hexadiene (mole ratio 85/8/7)

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Table 2

		Liquid crystalline polymer	Filler	Olefinic polymer	Liquid crystalline composition	Performance of box type molded product				
Kind	Amount (wt. part)	Kind	Amount (wt. part)	Kind	\overline{M}_w (wt. part)	Amount (wt. part)	Retention rate of weld strength (Z)	Hold releasing force (kg)	Change in shape of partition wall	Strength of weld
Example 5	A 100	Glass fiber	50	PE 4.0×10^3	0.45	27	25	None	3.7	
- 6	B 100	"	50	PE 4.0×10^3	0.45	26	25	-	3.6	
- 7	C 100	"	50	PE 4.0×10^3	0.45	20	32	-	2.5	
- 8	D 100	"	50	PE 4.0×10^3	0.45	18	32	-	2.5	
- 9	A 100	"	50	PE 4.0×10^3	0.05	28	45	-	3.8	
- 10	A 100	"	50	PE 4.0×10^3	1.50	25	22	-	3.5	
- 11	A 100	"	50	PE 3×10^4	0.45	26	49	-	3.5	
- 12	A 100	"	50	PP 1.5×10^3	0.45	26	30	-	3.4	
- 13	A 100	"	50	PE/P 1.0×10^3	0.45	26	31	-	3.5	
- 14	A 100	"	50	PE/B1 1.1×10^3	0.45	27	32	-	3.6	
- 15	A 100	"	50	PE/P/HD 8×10^4	0.45	25	35	-	3.4	
- 16	A 100	Glass fiber Hica	40	PE 4.0×10^3	0.45	23	26	-	3.1	
- 17	A 100	Glass fiber Talc	40	PE 4.0×10^3	0.45	23	27	-	2.9	
Comparative Ex.	A 100	Glass fiber	50	-	-	29	82	Yes	Cracking when released	
- 7	B 100	"	50	-	-	27	82	-	-	
- 8	C 100	"	50	-	-	21	85	-	-	
- 9	D 100	"	50	-	-	19	85	-	-	
- 10	A 100	"	50	PE 8000	0.45	10	69	-	0.7	
- 11	A 100	"	50	PE 3000	0.45	10	69	-	0.6	
- 12	A 100	"	50	PE 5000	0.45	9	69	-	0.6	

Examples 18-23

Sample pellets of compositions were produced in the same manner as Example 5, except that the organic bromine compounds listed in Table 4 were blended with liquid crystal polymer from the raw material feed port in the proportions indicated in Table 4 based upon 100 parts by weight of the liquid crystalline polymer in Example 1. These pellets were then fed into a Sumitomo Nestal Promat 40/25 Injection Molding Machine (Sumitomo Heavy Machine Industry K.K.) and molded into testpieces measuring 0.5 mm (thickness) × 12.7 mm × 127 mm, as well as strip testpieces measuring 0.8 mm (thickness) × 12.7 mm × 127 mm under conditions of a cylinder temperature of (the melting point of the liquid crystalline polymer + 10°C) and a mold temperature of 90°C. A vertical combustion test was then performed according to UL94 standards using the testpieces to evaluate flame retardency. The other evaluation tests were carried out in the same manner as in Example 5.

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Table 3

Liquid crystalline polymer		Filler		Olefinic polymer		Organic bromine compound		Reten- tion rate of weld strength	Performance of box type molded product	Mold releas- ing force (kg)	Change- in shape of parti- tion wall	Strength of weld (kg)	III.-9 Flame rotat- ency	
Kind	Amount	Kind	Amount	Kind ^{*1}	\overline{M}_w	Amount	Kind ^{*2}	Amount						
Example 18	A	100	Glass fiber	50	PE	4.0×10^5	0.45	FR-1	6	26	26	None	3.4	V-0
-	19	A	100	*	PE	4.0×10^5	0.45	FR-2	6	26	26	*	3.4	V-0
-	20	A	100	*	PE	4.0×10^5	0.45	FR-3	6	25	27	*	3.4	V-0
-	21	A	100	*	PE	4.0×10^5	0.45	FR-4	6	26	26	*	3.3	V-0
-	22	A	100	*	PE	4.0×10^5	0.45	FR-5	6	25	29	*	3.3	V-0
-	23	A	100	*	PE	4.0×10^5	0.45	FR-6	6	25	29	*	3.2	V-0

*1: PE - polyethylene
 *2: see Table 4

Table 4

No.	Structure
5	FR-1 Poly(brominated styrene) obtained by polymerization of a monomer containing 80% by weight of dibrominated styrene, 15% by weight of monobrominated styrene, and 5% by weight of tribrominated styrene (bromine content: 59%) Weight average molecular weight: 30×10^4
10	FR-2 Poly(brominated styrene) obtained by polymerization of tribrominated styrene monomer (bromine content: 68%) Weight average molecular weight: 30×10^4
15	FR-3 Dibrominated polystyrene obtained by bromination of polystyrene (bromine content: 60%) Weight average molecular weight: 26×10^4
20	FR-4 Tribrominated polystyrene obtained by bromination of polystyrene (bromine content: 68%) Weight average molecular weight: 25×10^4
25	FR-5 Brominated epoxy polymer
	FR-6 Brominated polycarbonate

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Examples 24-28

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Example 5 was further repeated except that carbon black was formulated as shown in Table 5 below.
The results are shown in Table 5.

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Table 5

Liquid crystalline polymer		Filler		Olefinic polymer		Carbon black		Retention rate of weld strength		Performance of box type molded product	
								Kind	Amount (wt. part)	Mold releasing force (kg)	Change in shape of partition wall
Example 24	A	100	Glass fiber	PE	4.0×10^3	0.45	7	0.2	26	26	None
25	A	100	*	PE	4.0×10^5	0.45	7	0.5	26	26	*
26	A	100	*	PE	4.0×10^3	0.45	7	1.0	25	26	*
27	A	100	*	PE	4.0×10^5	0.45	7	5.0	26	26	*
28	A	100	*	PE	4.0×10^3	0.45	5	1.0	25	26	*

*1: PE = polyethylene

As shown above, shaped articles of a liquid crystalline polymer composition embodying the present invention are excellent in thermal resistance, mechanical properties, dimensional stability and mold releasability and also excellent in the weld strength, and therefore, are suitable for various applications such as electrical and electronic equipment, precision machinery related equipment, office equipment, and automobile and vehicular related equipment.

Claims

- 10 1. A shaped article of a liquid crystalline polymer having a weld, which is provided by plural melted resin flows encountering in the mold, obtained by melt molding a composition comprising (A) 100 parts by weight of at least one liquid crystalline polymer selected from the group consisting of liquid crystalline polyesters and liquid crystalline polyester amides capable of forming an anisotropic melt and (B) 0.01 to 2 parts by weight of at least one olefinic polymer and having a weight-average molecular weight of 10,000 to 600,000 selected from the group consisting of (a) polyethylene, (b) polypropylene, (c) copolymers of ethylene and α -olefin having 3 or more carbon atoms, (d) copolymers of propylene and α -olefin having 4 or more carbon atoms, (e) copolymers of ethylene, α -olefin having 3 or more carbon atoms and a non-conjugated diene and (f) copolymers of propylene, α -olefin having 4 or more carbon atoms and a non-conjugated diene, said composition having a retention rate of a weld strength of 15% to 100% obtained by the following formula (1), based upon flexural strengths determined from a weld shaped article (X) having a weld, which is provided by plural melted resin flows encountering in the mold, at substantially the center portion thereof and a non-weld shaped article (Y) having no weld therein, both obtained by injection molding the composition under the following conditions:

$$\text{Retention rate of weld strength (\%)} = (\text{Flexural strength of shaped article (X) having weld}/\text{Flexural strength of shaped article (Y) having no weld}) \times 100 \quad (1)$$

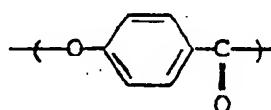
25 Injection molding condition:

Cylinder temperature: Melting point ($^{\circ}\text{C}$) of liquid crystalline polymer + 10 $^{\circ}\text{C}$

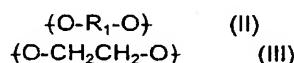
Mold temperature: 90 $^{\circ}\text{C}$

Size of shaped article: 6.0 mm (width) \times 127 mm (length) \times 1 mm (thickness).

- 30 2. A shaped article according to claim 1, having the weld portion with a retention rate of a weld strength is 20% to 100%.
- 35 3. A shaped article according to claim 1 or claim 2, wherein the liquid crystalline polymer (A) is at least one polymer selected from fully aromatic liquid crystalline polyesters, fully aromatic liquid crystalline polyesteramides, liquid crystalline polyesters having an ethylenedioxy unit, and liquid crystalline polyesteramides having an ethylenedioxy unit.
- 40 4. A shaped article according to claim 3, wherein the liquid crystalline polymer (A) is at least one polymer selected from liquid crystalline polyesters having an ethylenedioxy unit and liquid crystalline polyesteramides having an ethylenedioxy unit.
- 45 5. A shaped article according to claim 4, wherein the liquid crystalline polyester (A) comprises at least one liquid polyester components each said component containing a combination of structural units selected from (i)(I), (III) and (IV), (ii)(I), (II) and (IV) and (iii)(I), (II), (III) and (IV):



... (I)



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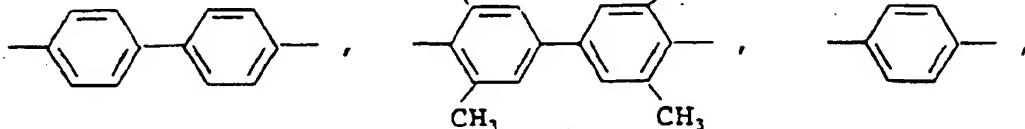
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wherein R₁ represents at least one group selected from:

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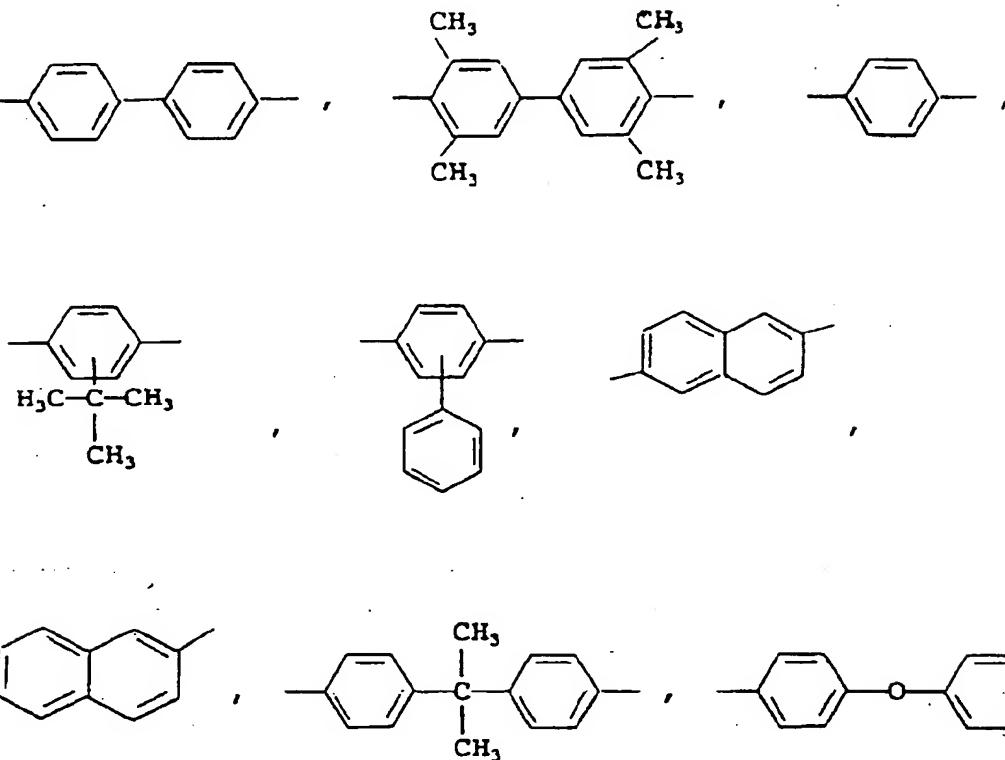
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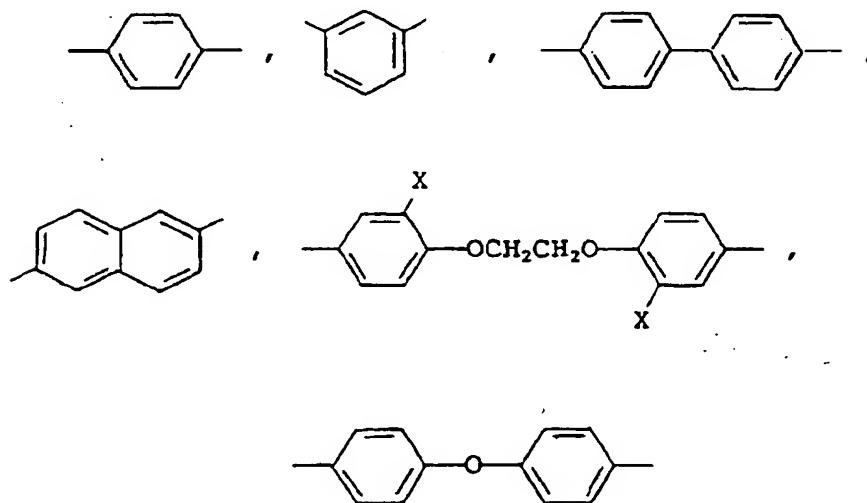


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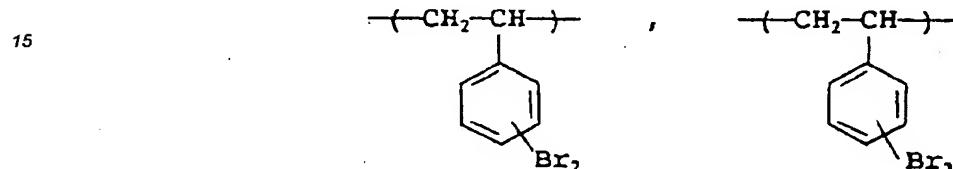
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wherein X represents a hydrogen atom or chlorine atom, and the total number of moles of structural unit

(II) and structural (III) are substantially equal to the total of the number of moles of structural unit (IV).

6. A shaped article according to any preceding claim, wherein the composition further contains 400 parts by weight or less of a filler, based upon 100 parts by weight of the liquid crystalline polymer (A).
- 5 7. A shaped article according to any preceding claim, wherein the composition further contains 0.5 to 60 parts by weight of an organic bromine compound based upon 100 parts by weight of the liquid crystalline polymer (A).
- 10 8. A shaped article according to claim 7, wherein the organic bromine compound is poly(brominated styrene) having a weight average molecular weight of 1×10^3 to 120×10^4 and having at least one of the following structural units obtained from brominated styrene monomer as the major structural component.



9. A shaped article according to any preceding claim, wherein the composition further contains 0.01 to 10 parts by weight of carbon black, based upon 100 parts by weight of the liquid crystalline polymer (A).
- 25 10. A shaped article formed by a moulding process in which at least two flowing molten resin portions unite during the molding to form a weld, which flowing molten resin portions are provided by a resin composition comprising (A) 100 parts by weight of at least one liquid crystalline polymer selected from the group consisting of liquid crystalline polyesters and liquid crystalline polyester amides capable of forming an anisotropic melt and (B) 0.01 to 2 parts by weight of at least one olefinic polymer and having a weight-average molecular weight of 10,000 to 600,000 selected from the group consisting of (a) polyethylene, (b) polypropylene, (c) copolymers of ethylene and α -olefin having 3 or more carbon atoms, (d) copolymers of propylene and α -olefin having 4 or more carbon atoms, (e) copolymers of ethylene, α -olefin having 3 or more carbon atoms and a non-conjugated diene and (f) copolymers of propylene, α -olefin having 4 or more carbon atoms and a non-conjugated diene.

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Fig.1

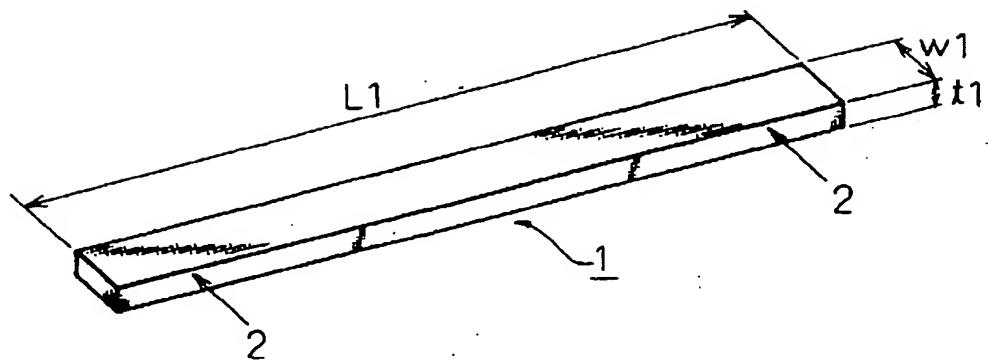
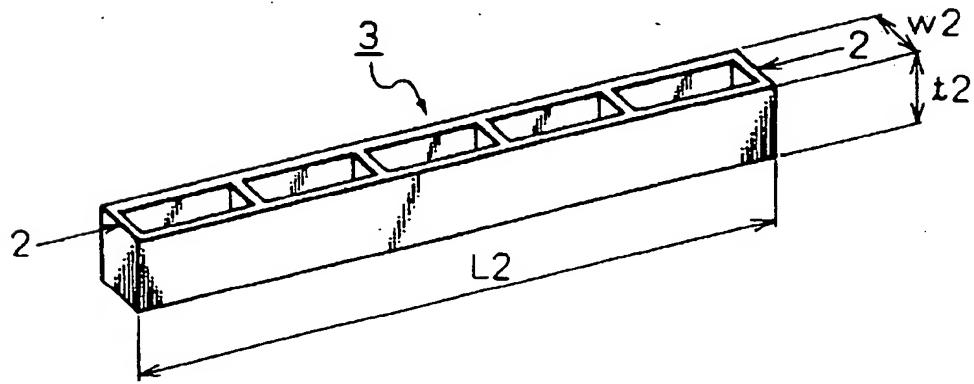


Fig. 2





(19) Europäisches Patentamt
European Patent Office
Office européen des brevets



(11) Publication number: 0 662 381 A3

(12)

EUROPEAN PATENT APPLICATION

(21) Application number: 94309859.0

(51) Int. Cl.⁶: B29C 45/00, C08L 67/00,
C08L 77/12, C09K 19/38,
// (C08L67/00, 23:02),
(C08L77/12, 23:02)

(22) Date of filing: 28.12.94

(30) Priority: 28.12.93 JP 338096/93

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12.07.95 Bulletin 95/28

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(84) Designated Contracting States:
DE FR GB

(88) Date of deferred publication of search report:
09.08.95 Bulletin 95/32

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(54) Shaped article of liquid crystalline polymer.

(57) A liquid crystalline polymer contains (A) 100 parts by weight of a liquid crystalline polyester and/or liquid crystal polyetheramide capable of forming an anisotropic melt and (B) 0.01 to 2 parts of an olefinic polymer. The composition is especially suitable for moulding therefrom shaped articles by a moulding process in which at least two flowing molten resin portions of the composition unite to form a weld. Thus, using such a composition it is possible to produce a moulding having such a weld in which the weld has a weld strength retention of 15% to 100%, so that the shaped article has excellent heat stability, mechanical properties, dimensional stability and mold releasability and is suitable for various application uses such as in electrical and electronic devices, accurate machine devices, office related devices and automobile parts.

EP 0 662 381 A3



European Patent
Office

EUROPEAN SEARCH REPORT

Application Number
EP 94 30 9859

DOCUMENTS CONSIDERED TO BE RELEVANT					
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)		
Y	EP-A-0 462 844 (TORAY INDUSTRIES, INC.) * abstract; claims 1-7, 13, 14 * * page 6, line 25 - line 46 * ---	1-11	B29C45/00 C08L67/00 C08L77/12 C09K19/38		
Y	EP-A-0 111 179 (GENERAL ELECTRIC COMP.) * abstract * * page 2, paragraph 2 - page 4, paragraph 1 * * page 10, paragraph 4 - page 12, paragraph 1 * ---	1-11	//(C08L67/00, 23:02), (C08L77/12, 23:02)		
X	EP-A-0 369 734 (AMOCO CORP.) * abstract; example 1 *	11			
X	EP-A-0 380 112 (KAWASAKI STEEL CORP.) * claims 1-3, 7; examples 12, 13, 22 * * page 2, line 4 - line 28 * * page 6, line 55 - page 7, line 16 * * page 17, line 1 - line 6 * ---	11			
X	PATENT ABSTRACTS OF JAPAN vol. 14, no. 74 (C-0687) 13 February 1990 & JP-A-01 292 057 (UENO SEIYAKU OYO KENKYUSHO KK) 24 November 1989 * abstract *	11	TECHNICAL FIELDS SEARCHED (Int.Cl.6) C08L C09K		
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CATEGORY OF CITED DOCUMENTS					
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document					
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Application Number
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